REPORT DOCUMENTATION PAGE

AFRL-SR-AR-TR-05-

0331

1. REPORT DATE *(DD-MM-YYYY)* 7/20/05	**2. REPORT TYPE** FINAL REPORT	**3. DATES COVERED** *(From - To)* September 1, 2001 - April 30, 2005

4. TITLE AND SUBTITLE
The Coherent Nonlinear Optical Response and Control of Single Quantum Dots

5a. CONTRACT NUMBER
F49620-01-1-0502

5b. GRANT NUMBER

5c. PROGRAM ELEMENT NUMBER

6. AUTHOR(S)
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5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
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8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
AFOSR
4015 Wilson Blvd. Room 713
Arilington, VA 22203-1954

NE

10. SPONSOR/MONITOR'S ACRONYM(S)
AFOSR

11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited

13. SUPPLEMENTARY NOTES

14. ABSTRACT
Work on this program is aimed at developing & understanding nano-optical structures with emphasis on developing quantum optical based devices. Specific work focused on semiconductor quantum dots. The major achievements include the first demonstration of an all optically driven quantum NOT-gate based on using the exciton-Bloch vector as the qubit (Science 2003). We also demonstrated optical density matrix tomography on this system. Since the fast recombination time of quantum dot excitons may limit these systems for quantum information applications, we initiated experiments on negatively charged quantum dots (similar to ions) where for quantum information processing, the qubit would be the electron spin. The energy level structure in this system is a 3-level - system, which is also of importance to novel coherent optical applications such as slow light & electromagnetic induced transparency. Our experiments in this system are just beginning, but we have successfully demonstrated optically induced coherence in this system, the unexpected effect of spontaneously generated spin coherence, & coherent optical control.

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF: a. REPORT	b. ABSTRACT	c. THIS PAGE	17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
U	U	U	UU	14	Duncan G. Steel
					19b. TELEPHONE NUMBER *(Include area code)* 734-764-4469

Standard Form 298 (Rev. 8/98)
Prescribed by ANSI Std. Z39.18

8-4-05

FINAL REPORT
To
THE AIR FORCE OFFICE OF SCIENTIFIC RESEARCH

The Coherent Nonlinear Optical Response and Control of Single Quantum Dots

AFOSR GRANT NO. F49620-01-1-0502

GRANT PERIOD: 9/1/01 – 4/30/05

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Abstract

Work on this program is aimed at developing and understanding nano-optical structures with emphasis on developing quantum optical based devices. Specific work focused on semiconductor quantum dots. The major achievements include the first demonstration of an all optically driven quantum NOT-gate based on using the exciton-Bloch vector as the qubit (*Science* 2003). We also demonstrated optical density matrix tomography on this system. Since the fast recombination time of quantum dot excitons may limit these systems for quantum information applications, we initiated experiments on negatively charged quantum dots (similar to ions) where for quantum information processing, the qubit would be the electron spin. The energy level structure in this system is a 3-level Λ-system, which is also of importance to novel coherent optical applications such as slow light and electromagnetic induced transparency. Our experiments in this system are just beginning, but we have successfully demonstrated optically induced coherence in this system, the unexpected effect of spontaneously generated spin coherence, and coherent optical control. A summary of our work leading up to this was published by invitation in *Physics Today* while the achievement of the quantum NOT-gate was reviewed in *Photonics News.*

PUBLICATIONS

(The most important reprints are included as an appendix)

JOURNAL PUBLICATIONS

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58. Jun Cheng, Xiaodong Xu, and D. G. Steel, A. S. Bracker and D. Gammon, L. J. Sham, “Spin-coherence-induced Hole Burning in Charged GaAs Quantum Dots,” QELS/CLEO 2005.

EDUCATIONAL ACTIVITY

A number of students participated in the program as evidenced in the above publications. Six of the students have since graduated with a Ph.D and gone on to postdocs or permanent positions including one student taking a permanent position at Lucent and another student joining NRL. Several new students have joined the group and are involved in the new program.

COLLABORATIONS

The work in the program is the result of an intense collaboration with Dr. D. Gammon at The Naval Research Laboratory supported by DARPA to develop quantum dot structures and spintronic based devices. In addition, the manybody theory component of the analysis of our findings is supported through our collaboration with Professor L.J. Sham (UCSD), supported by ARO, AFOSR and NSF.

SUMMARY OF FINDINGS

Nearly all of the research findings presented in this report have been reported in the annual reports. However, for completeness, we list the major developments, and we review a few of the most important results.

Introduction

The objectives of this program focused on developing and applying quantum optical methods based on coherent nonlinear laser spectroscopy to the study of physics in nanoscopic semiconductor structures. In the nanoscopic limit, we are focusing on semiconductor quantum dots, their physics, optical properties and the manifestations of inter-dot interactions. Quantum dots feature prominently in recent work to build photon-on-demand optical sources, low noise detectors, quantum information science, electromagnetic induced transparency (EIT) based devices, and spintronics. In this program, our work was aimed at studies of the basic physics in these systems, but, as in the case of our recent publication in Science demonstrating a quantum C-NOT, has a clear relationship to optically driven quantum computing.

The experimental approach is based primarily on various near field optical techniques, including both apertured and unapertured GaAs/AlGaAs epitaxially grown quantum dot structures.

Summary of the most important achievements:

Demonstration of all optical quantum Controlled-NOT gate.

Demonstration of optical modulation spectroscopy of exciton absorption in a single quantum dot.

Direct measurement of linear absorption from a single quantum dot.

Measurement of the biexciton coherence in an interface dot.

Detection of the coherent nonlinear optical response from excited states in a single dot.

Measurement of the biexciton coherence with the ground state in a self-assembled dot.

Observation of Raman coherence beats between orthogonally polarized excitons in a neutral quantum dot.

Demonstration of quantum entanglements between quantum states of adjacent dots.

Measurement of exciton Raman decoherence rate from quantum beats in self-assembled dots.

Demonstration of density matrix tomography on a single quantum dot.

Demonstration of quantum spin coherence and measurement of the ensemble spin-decoherence time.

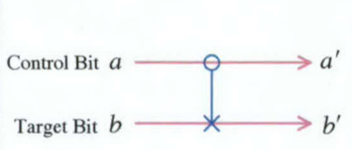
Demonstration of a viable 3-level lambda system in a semiconductor for applications to EIT type problems.

Demonstration of the coherent optical control of the spin-coherence.

Demonstration of an all Optically Driven Quantum Dot Controlled-NOT gate

While we are now moving on to work on single spin qubits to develop a quantum gate of the type first described by our collaborators and us in a recent Phys. Rev. Lett., we are completing the demonstration experiments of quantum operations using exciton qubits to demonstrate that manybody body effects that characterize the fast decoherence and interfere with coherent optical control in higher dimensional systems are suppressed in quantum dots leading to more atomic like behavior.

In this program, we succeeded in achieving the basic demonstration needed for proving feasibility of an optically driven solid state quantum computer. Specifically, we demonstrated coherent optical control of the quantum motion of one or two electrons in effective isolation in a semiconductor nanosystem leading to *the first demonstration of an all optically controlled semiconductor quantum NOT-gate with optical readout.* The result, published in Science (2003) demonstrated for the first time that semiconductors could provide a viable system for developing advanced ideas in quantum information science.



a	b	a'	b'
0	0	0	0
0	1	0	1
1	0	1	1
1	1	1	0

Figure 1. The controlled NOT-Gate and corresponding truth table. 'a' is the value of the control bit and 'b' is the value of the target bit for the conditional gate. .

A controlled-NOT gate is schematically illustrated in Fig. 1. A target bit may or may not change its state depending on the state of a control bit. The truth table for a controlled-NOT is shown on the right.

In a neutral quantum dot, two exciton optical Bloch vectors represent the two qubits. The Bloch vectors are described by orthogonally polarized excitation dipoles. As discussed in our previous Final Report, we had already demonstrated that Coulomb coupling between these two excitonic transitions leading to the biexciton produced the necessary interaction to enable quantum entanglement. The four qubits are $|00\rangle, |01\rangle, |10\rangle, and |11\rangle$ corresponding to no excitons present, 1 $\sigma+$ polarized exciton, 1 $\sigma-$ polarized exciton and two excitons corresponding to the biexciton, respectively.

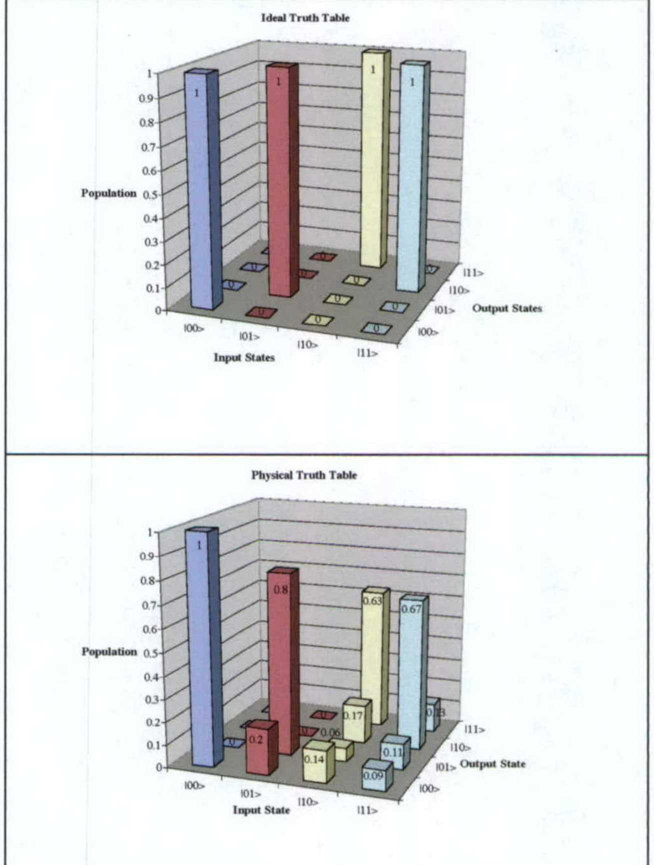


Figure 2. Experimentally determined truth table for a quantum controlled NOT gate. *Science*. **301**, 809 (2003). The upper panel is the ideal result, the bottom panel is the experimental value.

The pulse control of the biexciton dynamics, combined with the demonstrated control of the single exciton Rabi rotation, serves as the physical basis for a two-bit conditional quantum logic gate. The experimentally determined truth table for the gate (see Fig. 2 which shows the magnitude of the density matrix terms based on the experiments) shows the features of an all-optical quantum gate using interacting, yet distinguishable exciton transitions. Based on various optically initialized starting states, evaluation of the fidelity yields a calculated value of order 0.7 for the gate operation. This logic gate is the basis for further development of inter-dot logic gates for universal quantum computation.

Coherent Optical Tomography of Neutral Quantum Dot Excitation

Coherent control enabling an arbitrary rotation of the optical Bloch spin vector associated with either the optical dipole or the electron spin is an essential step in demonstrating coherent optical control of any electronic system for many proposed applications, including quantum computing. In the case of quantum computing, for example, single qubit (Bloch vector) rotations are often used to prepare the input states for logic operations. Fully characterizing the density matrix associated with this rotation is density matrix tomography. Alternatively, the performance of a quantum logical gate, such as a controlled-NOT gate in a semiconductor quantum dot, is characterized by its gate fidelity. In order to calculate the realistic gate fidelity of a particular gate, one must have full knowledge of the output state created by the gate operation, namely all values of the complex density matrix must be measured. Here we demonstrate the ability to measure the off-diagonal matrix elements (ρ_{12}, ρ_{21}) as well as the diagonal density matrix elements (ρ_{11}, ρ_{22}) of a closed two-level system, which leads to a complete mapping of the density matrix of a given qubit in a single quantum dot (QD).

This requires new capability for the laser system, so our objective in this was to use the exciton system (rather than spin), which is more established, and demonstrate that the new optical capability can achieve this new result.

In the exciton regime, optical excitation driving transitions between the crystal ground state and the exciton state translates to a rotation of the Bloch vector or qubit from state |0> to |1>. The optical pulse area, θ, is defined by $\theta(t)=\left[\mu_{eg}/\hbar\right]\int_{-\infty}^{\infty} dtE(t)$. A pulse area of π exchanges the two basis states, |0> and |1>, of the qubit, in particular, the populations between these states, i.e., population in qubit state |0> will be fully inverted to state |1> or vice versa. In general, we can apply pulses with corresponding pulse areas from 0 to π to create from the ground state (say |0>) an arbitrary superposition of |0> and |1>. This ability is demonstrated in the optically driven single exciton Rabi oscillations

Rabi oscillations manifest themselves in the fact that the population of the exciton population can be controlled coherently with a single laser pulse. However, multiple consecutive rotations of a qubit are required in practical quantum computation processes or to reconstruct the density matrix. Maintaining the relative phase information between consecutive single rotations is essential for these applications. E.g., the Hadamard gates, similar to an excitation with a π/2 pulse, that are used to prepare and collapse the qubit in logic operations in error correction codes and in the DJ problem, need to have a strict phase relation with each other in order to achieve the correct output.

Coherent optical studies show that the phase coherence of an exciton qubit is of the order of the exciton lifetime, implying negligible pure dephasing. In other proposed quantum systems such as nuclear magnetic resonance (NMR) and trapped ions where microsecond wide pulses are used, phase locking between rotations can be done electronically. In the QD systems, the THz computational speed is beyond the phase locking capability of even the fastest electronics available.

To maintain the phase coherence between rotations without compromising the speed of computation, an optical approach to phase locking is used. An experiment demonstrating the capacity of optical phase locking is conducted using two actively phase-locked pulses from a Michelson interferometer on an exciton-based qubit. The delay and the phase between the two pump pulses can be adjusted with a translation stage and a piezoelectric mount, respectively. The piezoelectric mount is controlled by a feedback loop to compensate for phase jitter in the optical system.

The experiment is conducted with a 4 ps pulse width laser in a pump and probe geometry. A Michelson interferometer in the path of the pump beam creates two pump pulses (P_1, P_2) with tunable coarse (τ) and phase (ϕ) delays between the pulses. The differential transmission (DT) signal is homodyne detected with the transmitted probe beam. The first pump pulse (P_1) with an arbitrary pulse area ($P_1\theta$) creates a state with an arbitrary density matrix,

$$M1 = \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{bmatrix} \qquad (1)$$

We can measure ρ_{22} with a simple population readout via DT which has a definite relation with the excited state population, then ρ_{11} is inferred from $1-\rho_{22}$. The second pump pulse (P_2) with a pulse area ($P_{2\theta}$) performs a rotation ($R_{P2\theta}$) to a new state characterized by a new density matrix,

$$M2 = \begin{bmatrix} \rho'_{11} & \rho'_{12} \\ \rho'_{21} & \rho'_{22} \end{bmatrix} = R_{P_{2\theta}} M1 R^{+}_{P_{2\theta}} \qquad (2)$$

For $P_{2\theta} = \pi/2$, $\rho'_{22} = 1/2 + \mathrm{Im}(\rho_{12})$ at $\phi = 0$ and $\rho'_{22} = 1/2 - \mathrm{Re}(\rho_{12})$ at $\phi = \pi/2$ for a closed two-level system. Since we can measure ρ'_{22} through population readout, we can extract the values of ρ_{12} and ρ_{21}. In the Bloch picture, the second pump pulse (P_2) corresponds to an active rotation of the Bloch state vector on the Bloch sphere, which projects the off-diagonal components of the vector onto the diagonal component axis of the Bloch sphere, making them extractable through population readout. This active $\pi/2$ rotation technique is widely used with magnetic fields in nuclear magnetic resonance (NMR). Here the method is extended to optical fields in a semiconductor system.

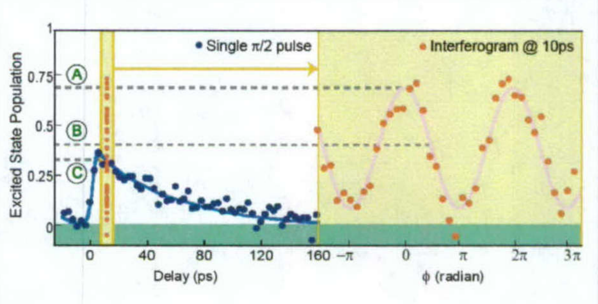


Fig. 3. Decay plots of a single $\pi/2$ excitation and an interferogram taken at τ=10 ps delay. The horizontal dash lines mark the value of each population readout. A gives ρ'_{22}= 0.73, B gives ρ'_{22}= 0.4 and C gives ρ_{22}= 0.35.

Using equation 2 and values A, B and C from Figure 3, we obtained ρ_{11}=0.65, ρ_{22}=0.35, $\rho_{12}=\rho^*_{12}$=0.11+i0.21 at τ=10 ps. In a simulation calculation, the values of

ρ_{11}= 0.60; ρ_{22}= 0.40, and $\rho_{12}=\rho^*_{12}$=0.08+i0.21 are comparable to the measured values. Due to the finite population decay and dephasing of this two-level system, there exist discrepancies between the decoherence-free values (ρ_{11}= 0.5, ρ_{22}= 0.5, $\rho_{12}=\rho^*_{12}$= i0.5) and the measured values of the density matrix elements created by P_1. Shorter pulse widths and systems with longer decay times will eliminate these discrepancies.

In an ideal Bloch sphere representation (Fig. 4), the qubit state prepared in (|0> + |1>)/2 is driven to the pseudo-spin down state for $\phi = \pi$ (Fig. 4a) and to the pseudo-spin up state for $\phi = 2\pi$ (Fig. 4b). In reality, the experimental results deviate from the ideal.

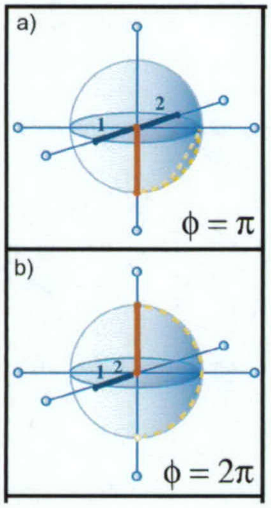


Figure 4. The phase dependent rotation of the Bloch spin vector in the exciton system. a) and b) are the rotations in the Bloch sphere picture. The red bars represent the state vectors and the blue bars represent the field vectors. The numbers denote the pump pulses 1 and 2.

Even though the lifetime of the qubit given the pulse width used is inadequate for the demonstration of a complete algorithm, it is enough to illustrate another important point: the quantum memory of the system. Due to the memory of the system, the exciton still 'remembers' the information imposed by the first pulse beyond the pulse duration, enabling the second pulse to access this information at a later time. The interferogram in Fig. 3 presents a clear example of the quantum memory, which allows two pulses to interfere with each other even when they are not overlapped in time. The span of the memory and the coherence lifetime of the system are essentially equivalent. For practical computation, a system with long coherence time, such as the spin system, is needed.

Interdot Quantum Entanglement

For many types of quantum information and quantum device applications, it is important for quantum systems to couple and preserve quantum coherence. For quantum computing, it must be possible to entangle quantum states in separate quantum dots. Figure 5 shows a cartoon illustrating the idea that if adjacent dots are sufficiently close to each other, it will be possible

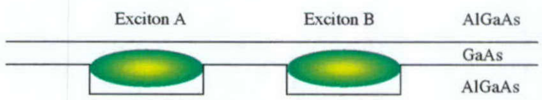


Figure. 5 A cartoon depicting two adjacent dots. If they are sufficiently close it will enable an interaction (e.g., dipole-dipole coupling) between the two localized excitons that can lead to quantum entanglement as illustrated by the data in Fig. 6.

In an earlier Final Report, we demonstrated coupling between dots leading to energy relaxation. However, this coupling was completely incoherent, and there was no evidence of coherent coupling. In this program, we have worked on developing the theory of coherent nonlinear optical spectroscopy of coupled systems in order to understand what the signatures would look like.

Based on these results, we were able to examine the interface fluctuation dot structures and find examples of coherently coupled quantum dots. Figure 6 shows an example of the results demonstrating interdot quantum entanglement.

For these experiments two cw optical laser fields with a mutual bandwidth of order 100 neV are arranged in the pump-and-probe geometry nearly parallel to the B-field, and the coherent nonlinear signal is homodyne-detected with the transmitted beams. The fields are either co- or cross-circularly polarized to excite and probe well-defined dipole transitions.

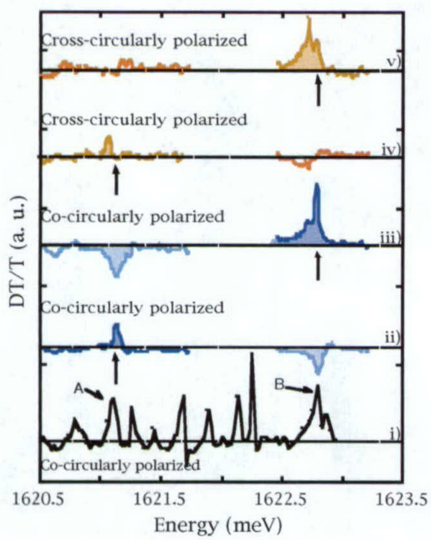


Figure 6: Differential transmission (DT) spectra performed with an applied magnetic field of 0.5T in the Faraday configuration. i) Degenerate DT with excitons of interest labeled A and B. When peak A (B) is excited, saturation of the exciton is seen at resonance, but induced absorption is seen at B (A) when the beams are co-circularly polarized (ii-iii). If the beams are cross-circularly polarized (iv-v), no induced absorption is seen.

The degenerate DT/T spectrum in Figure 6 maps out the spectrum of heavy hole excitons. The two spectral lines of interest are at 1621.13 meV (A) and 1622.80 meV (B). The non-degenerate spectra in Figure 6 (ii-iii) is obtained by placing the pump beam at resonance with one of the spectral lines and scanning the probe beam over the complete spectrum. When the pump is at A (B) and the probe is co-polarized, the exciton resonance shows saturation at A (B) as expected. However, at B (A), induced absorption is seen.

If the two beams are cross-circularly polarized (iv-v in Figure 6), the induced absorption dip is not seen. In dots that exhibit this structure (about 3%), no bound state biexcitons (typically with binding energy, Δ=-3.5 meV) are observed.

The phenomenological Hamiltonian that predicts the spectral features seen in this system and the absence of the single dot biexciton is a combination of two interacting two-exciton systems and gives a theoretical prediction very similar to that observed. These results serve now as the basis for designing our future experiments to study interacting quantum dot spin systems, discussed below.

Coherent Optical Excitation and Readout of Electron Spin Coherence in Charged Quantum Dots

By adding a single electron to a quantum dot, we convert the neutral quantum dot to a charged dot where the ground state is doubly degenerate. This system leads to the basic 3-level Λ excitation scheme critical to so many proposed applications and devices. In reality, the system is slightly more complex because there are two upper level states (called a charged exciton or trion) that contribute to the overall optical response. This system features prominently in proposals for quantum computing since the two degenerate ground states (in the case of no magnetic field) correspond to two orthogonal spin states of the electron. This system also features in proposals for solid state based systems for applications of electromagnetic induced transparency. This system is of interest to both of these areas of research since quantum spin coherence should have long decoherence times (>>100 microseconds).

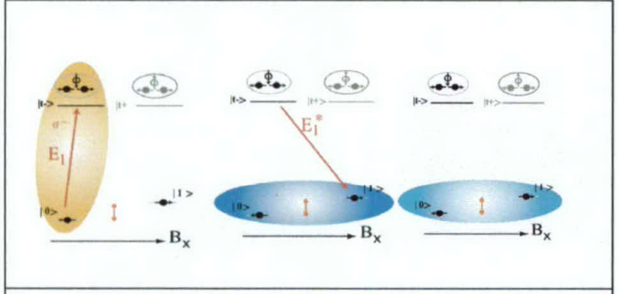


Figure 7. This diagram illustrates how two optical pulses can lead to quantum coherence. The shaded region identifies the optically induced coherence following excitation by the given field.

In this program, we initiated measurements to demonstrate and understand optically induced and detected electron spin coherence. In these measurements, we used a fully resonant Raman excitation scheme, shown in Fig. 7. A pulsed optical field with a bandwidth large compared to the Zeeman splitting of the spin states is tuned to the trion transition. The field then excites the Raman coherence, i.e., creates a coherent superposition of spin up and spin down, aligned along the magnetic field direction, x. This coherence oscillates in time at the Zeeman splitting energy. A probe field samples the coherence as a function of time after the pump excitation, and the coherent optical emission stimulated by the probe is then homodyne detected with the probe field. The spin oscillation is shown in Fig. 7.

Two sets of data are shown in Fig. 8. The lower curves on the left and right are data obtained from an ensemble of quantum dots. These represent our first data obtained in this system. The data show the distinct oscillations of a true quantum beat between the spin states. While the life time of this coherence is anticipated to be very long, there is a decay of the envelope that reflects a decoherence time of 10 nsec in the limit that B goes to zero (see Fig. 9). The decay is believed to arise from inhomogeneous broadening of the Zeeman splitting due to variations in the g-factor.

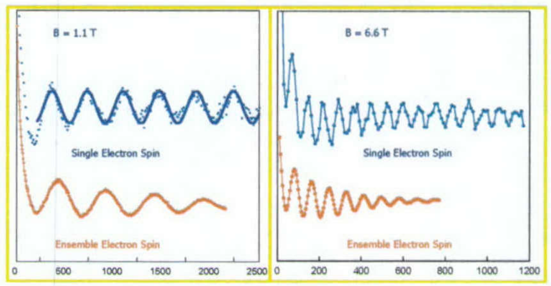


Figure 8. Experimental data showing quantum beats arising from optically induced electron spin coherence. Data in the left and right panels correspond to two different magnetic fields. The red curves represent data obtained on an ensemble of single electron doped quantum dots. The decay of the beats arises from inhomogeneous broadening, most likely of the electron g-factors. The blue curves show the quantum beats obtained from a single quantum dot. The data on left panel shows no detectable decay. The data on the right shows that the apparent existence of sudden unexplained changes in beat amplitude.

More recent measurements have now been achieved on single quantum dots. The upper curves in Fig. 8 show this newest data. A careful fit of the data shows no detectable decay, as expected. However, there remain many aspects of the data that are under investigation.

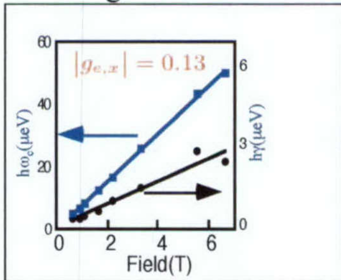


Figure 9. Data obtained from ensemble results given the splitting and decoherence rate as function of applied magnetic field. At B=0, the ensemble decoherence time is 10 nsec.

Careful inspection of the data in Fig. 8 shows an anomalous chirp in the oscillation. Also, there appear to be jumps in the amplitude of the data at long times (on the right), which suggest some kind of change in the dot. New studies that seek to eliminate the oscillation in the measurement and just measure the decay are expected to eliminate some of the issues.

Spontaneously generated coherence

In the quantum beat data above, an anomalous variation in the beat amplitude and phase occurred as a function of splitting of the two ground states. In the standard theory for a 3-level Λ system, there is no change in either parameter, as illustrated by the dashed lines in Fig. 10. However, the data clearly shows a dependence on splitting that only approaches the standard result for large splitting.

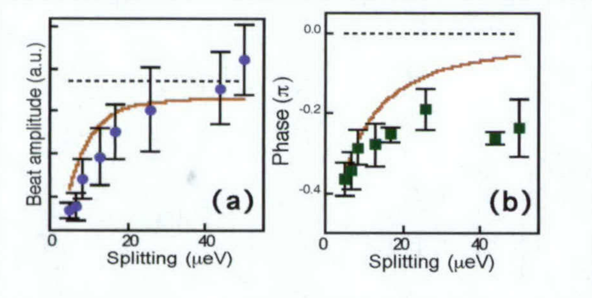


Figure 10. The quantum beat amplitude and phase of Fig. 8 shows an anomalous dependence on splitting. The dashed line is the standard theory. The solid line is the theory that accounts for decay of the trion state to a quantum coherence between the ground states (spontaneously generated coherence).

The origin of this behavior is in fact due to completely new physics not observed in atomic systems. Specifically, the theory shows that this dependence on splitting is due to spontaneously generated coherence (SGC). As illustrated in Fig. 11, in general, an excited state can decay simultaneously to both ground states giving rise to a quantum coherence. This can occur if the dipole moments associated with these transitions are not completely orthogonal, and physically corresponds to an interference that results when the two quantum mechanical decay pathways can not be distinguished. In atomic systems, this is forbidden by symmetry considerations. However, in these quantum dots, the axis of quantization remains pinned in the growth direction, and hence application of a magnetic field in the x-direction breaks this symmetry, and destroys the polarization selection rules.

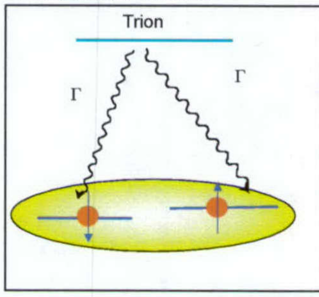


Figure 11. Energy level diagram for 3-evel system where the excited state is allowed to decay to the ground state with non-orthogonal dipole moments.

Coherent Optical Control of Electron Spin Coherence

In our most recent work, not yet published, we have demonstrated coherent optical control of the spin coherence. These results are a major advance in the ability to optically manipulate electron spins and build on our earlier work demonstrating coherent optical control of the exciton Bloch vector (Science, 1998).

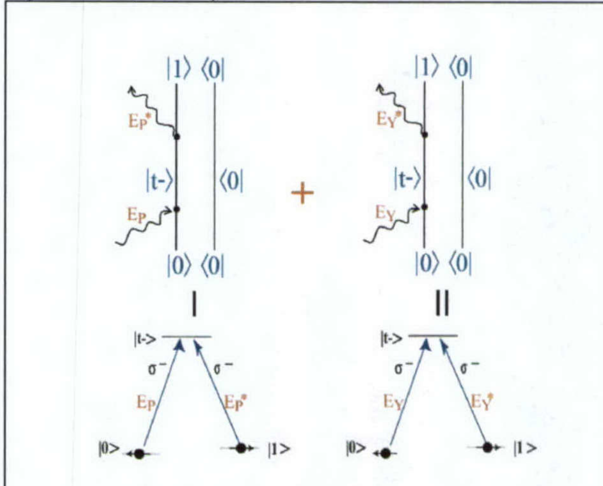


Figure 12. Double side Feynman diagrams showing the origin of coherent optical control of spin. This control is achieved by the interference of two optically excited pathways leading to production of spin coherence between the two spin states.

The physical basis for optical control of spin coherence is shown in Fig. 12. Here, two double-sided Feynman diagrams represent two pathways to formation of spin coherence between the two spin states that interfere depending on the phase difference between the two excitation paths. By adjusting the phase difference, the interference can be either constructive or destructive.

Figure 13 shows the experimental result. At t=0 the first excitation pulse creates electronic spin coherence. At a given phase delay (given by the ratio of the time delay between the excitation pulse to the beat period) a second pulse controls the coherence by constructive or destructive interference. In the usual way (see Fig 8), the probe pulse then follows the time evolution of the quantum coherence.

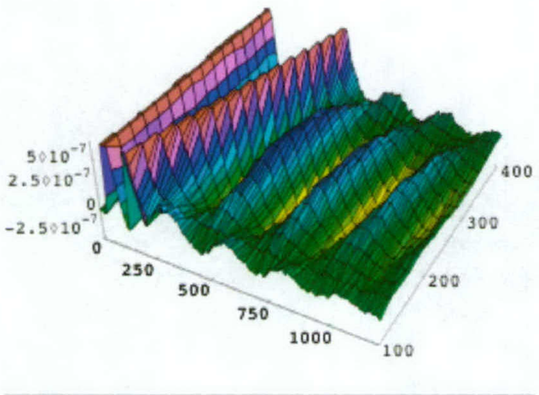


Figure 13. Experimental results demonstrating coherent optical control of electron spin coherence. The x-axis represent the time evolution of the quantum coherence showing quantum beats similar to Fig. 8. The y-axis represent the phase delay (in time) between the spin coherence forming pulse and the control pulse. Constructive interference is seen around a phase delay of 300.

Summary

This program has resulted in new understanding of importance to nonlinear optical nano-science, and has shown that nano-structures may be potentially viable structures for optically controlled quantum devices. The studies have now evolved to studies of charged quantum dots that give rise to an energy level structure similar to that needed for many innovative applications such as for creating slow light devices and quantum logic gates. This work has laid the foundation for our future studies of spin based quantum dots and more complex structures based on interacting quantum systems.